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On the Analysis of $Cd_2Nb_2O_7$ Dielectric Dispersion

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Abstract

The dispersion data of $Cd_2Nb_2O_7$ (CNO) ceramics have been analyzed using multiple-arc methodology. The high frequency arc has revealed a Ferroelectric property for the ceramics. Also, the method has indicated that mere Cole-Cole single arc analysis over a limited part of the dispersion data may not lead to a reliable dispersion parameters.

Keywords: Dielectric; Ferroelectric; Ceramics.

1. Introduction

Various practical techniques and numerous theoretical approaches have been employed or developed in dielectric analysis [1-3]. This may reflects the complexity of dielectric system in general. Impedance measurements are among effective methods adopted in investigating the dispersion in dielectrics as a means to explore the underlying mechanisms governing the dielectric response. Cole-Cole plots are commonly employed in the analysis of the impedance data under different conditions such as temperature; and that is due to its virtue of being simple to implement. The associated relations of the plot exhibit a circular arc in the complex plane of dielectric constant, $\varepsilon=\varepsilon'-j\varepsilon''$. The important parameters extracted from the plot are the spread in the relaxation time and its mean value which are the common features in majority of dielectrics characterized by deviations from the Debye single relaxation in dispersion. However, the frequent use of the method in the research and duly reported in literatures is confined to a limited frequency range of the measured data where a single-arc is definable; thereby leaving the

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remaining data unattainable by the analysis [4]. In some cases, the conclusions drawn from the Cole-Cole approach render them questionable in their validity.
Alternatively, multiple-arc method has the potential in covering the entire data range used. The approach is simple to implement in the sense that it is derived as an extension of the Cole-Cole method. The comprehensive coverage of data is expressed essentially by the relaxation time distribution, RTD, which takes the form, [5]

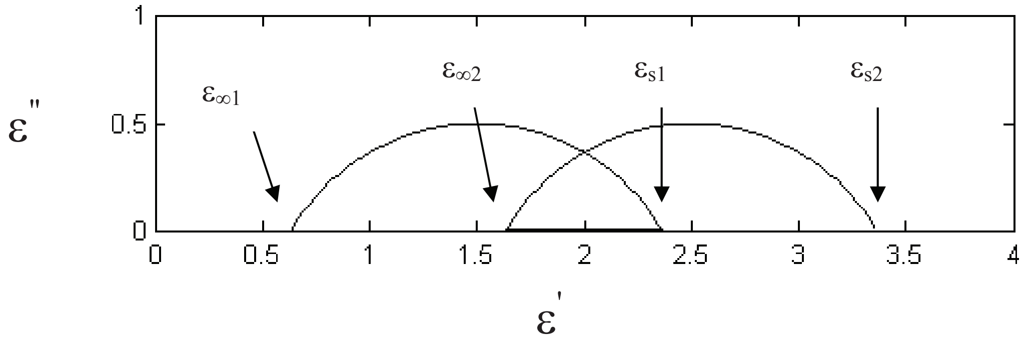

Fig.(1): The (ε''-ε') relation illustrating two overlapping arcs.

$$\mathrm{f(u)} = \mathrm{C}\sum_{\mathrm{n=1}}^{\mathrm{m}} \frac{(\varepsilon_{sn} - \varepsilon_{\infty n})\sin(\alpha_n \pi)}{Cosh[(1-\alpha_n)(u+\beta_n)] - \cos(\alpha_n \pi)} \quad , \tag{1}$$

where $\mathrm{C} = [2\pi \sum (\varepsilon_{\mathrm{sn}} - \varepsilon_{\infty n})]^{-1}$, $\beta_{\mathrm{n}} = \ln(\tau_{01} / \tau_{on})$, $\mathrm{u} = \ln(\tau / \tau_{01})$, with $\int_{-\infty}^{\infty} \mathrm{f(u)du} = 1$.
The corresponding expression for ε' and ε'' are given in terms of the RTD as,

$$\varepsilon'(\omega) = \varepsilon_\infty + \sum_{\mathrm{n=1}}^{\mathrm{m}} (\varepsilon_{\mathrm{sn}} - \varepsilon_{\infty n}) \int_{-\infty}^{\infty} \frac{\mathrm{f(u)}}{1+\omega^2\tau^2}\mathrm{du} - \varepsilon_c(\omega) \tag{2}$$

and

$$\varepsilon''(\omega) = \sum_{\mathrm{n=1}}^{\mathrm{m}} (\varepsilon_{\mathrm{sn}} - \varepsilon_{\infty n}) \int_{-\infty}^{\infty} \frac{\tau\omega \mathrm{f(u)}}{1+\omega^2\tau^2}\mathrm{du} \tag{3}$$

$\varepsilon_{\mathrm{sn}}$ and $\varepsilon_{\infty\mathrm{n}}$ are the low- and high-frequency dielectric constants, illustrated in Fig.(1), of the n^{th} arc respectively, while m designates the number of arcs traced over the frequency range in use with α_{n} as the spread parameter for the n^{th} arc taking values between 0 and 1. τ_{on} represents the most probable relaxation time for the n^{th} arc, while τ_{o1} represents an arbitrary chosen low frequency reference arc. The $\varepsilon_c(\omega)$ term is a correction function ascribed to the arcs' overlapping, depicted in Fig.(1) [6,7]. The known Cole-Cole formula is then regarded as a special case of equation (1) where n=1. Each term in the distribution above corresponds to a single-arc distribution multiplied by the weighting factor,

$$(\varepsilon_{\mathrm{sn}} - \varepsilon_{\infty n})\left[\sum_{\mathrm{n=1}}^{\mathrm{m}} (\varepsilon_{\mathrm{sn}} - \varepsilon_{\infty n})\right]^{-1}$$

As the distribution in (1) represents a statistical average for different single-arc distributions, it, presumably, reflects the relative effect of various categories of polarization occurring in a material subjected to varying frequency and temperature. It is worth noting in Fig.(1) that arcs may overlap pronouncly and the main factor specifying the overlapping degree is the relative order of magnitude of the relaxation time τ_o between arcs. The overlapping is also reflected in the spread parameter α**.** For instance, weak arcs overlapping is experienced in KNbO3 [8] and ZnO [9] materials. In such cases analysis of arcs become almost independent. This letter encounters the dispersion analysis of $Cd_2Nb_2O_7$ (CNO) Ferroelectric ceramics reported by Chen Ang et al. [10].

1. The dispersion of (CNO) ceramics:

Three sets of data are given in the reference for ε' and ε'' at different temperatures over the frequency range 10^2-10^5 Hz. Their sets of data were each fitted to a single arc in the ε'-ε'' plane and four parameters were then evaluated from each plot. However, the fits were limited to parts of the frequency range while significant part of the remaining data points deviates from the Cole-Cole arcs and hence become incompliant with single arc approach. Subsequently, the main parameters extracted from the fits and then used in the analysis were the relaxation times for the three sets. Parameters such as activation energy for a particular relaxation process were then derived through Arrhenius plots and augmented by the empirical Vogal-Fulcher relation [10].

2. Arcs' analysis of CNO ceramics:

In our approach the three sets of data were subjected to the analysis by the multiple-arc method. The validation of arc analysis in this context was examined by comparing the ε'-ω and ε''-ω with the experimental data. The fitting procedure followed the steps outlined in ref. [7] whereby the high frequency data is first treated. The same procedure is then followed with the remaining data. Within the frequency range reported the data for each temperature were fitted to two arcs and the corresponding parameters are given in table I.

It is of significance to find that the high frequency arc undergoes a decrease in the spread parameter α from 0.7 to 0.28 while the temperature changes from 75 to 180 K. This feature resembles that one found for KNbO3 ceramic which was then attributed to Ferroelctric property [8]. In addition, the large α values indicate strong overlapping between arcs for the three temperatures. The relatively large difference between ε_s of the high frequency arc and ε_∞ of the low frequency arc is another indication of strong overlapping which masked the circular feature of the arcs. Fig.(2) depicts satisfactory agreement between experimental and theoretically evaluated ε'-ε'' relations over substantial part of the range. The deviations from the fits at the lower and upper parts of the frequency range are likely to be ascribed to other traceable arcs if sufficient data were availed in these ranges. Using equations 1, 2 & 3 along with the parameters in table I, Fig.(3) and Fig.(4) show satisfactory agreements achieved with reported experimental data within an error of no more than 10%.

Table I: Arcs Dispersion Parameters for CNO Ferroelectric Ceramics

T° K	Arc I (Low frequency)				Arc II (High frequency)			
	ε_s	ε_∞	α	τ_o (*ms*)	ε_s	ε_∞	α	τ_o (μS)
75	**1261**	**1156**	**0.6**	**1.5**	**1228**	**1078**	**0.7**	**10.0**
126	**1780**	**1543**	**0.41**	**1.5**	**1630**	**1383**	**0.5**	**16.0**
180	**4861**	**4360**	**0.52**	**0.25**	**4505**	**4232**	**0.28**	**5.0**

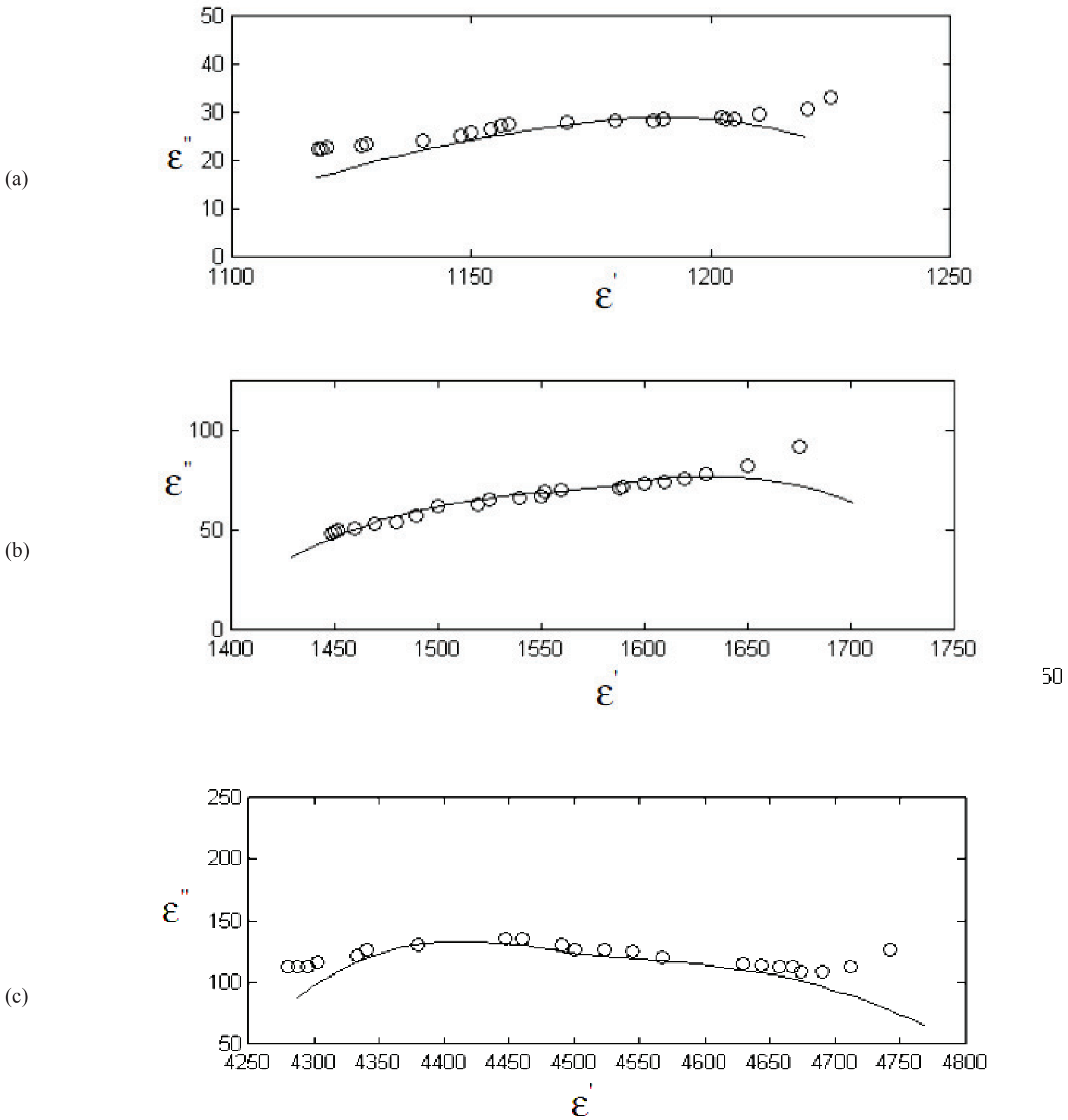

Fig.(2): The (ε''-ε') relation for CNO ceramic in the frequency interval 10^2-10^5 HZ

at (a) 75 K temperature, (b) 126 K temperature and (b) 180 K temperature

.__: Theoretical fit to two arcs

O: experimental data points

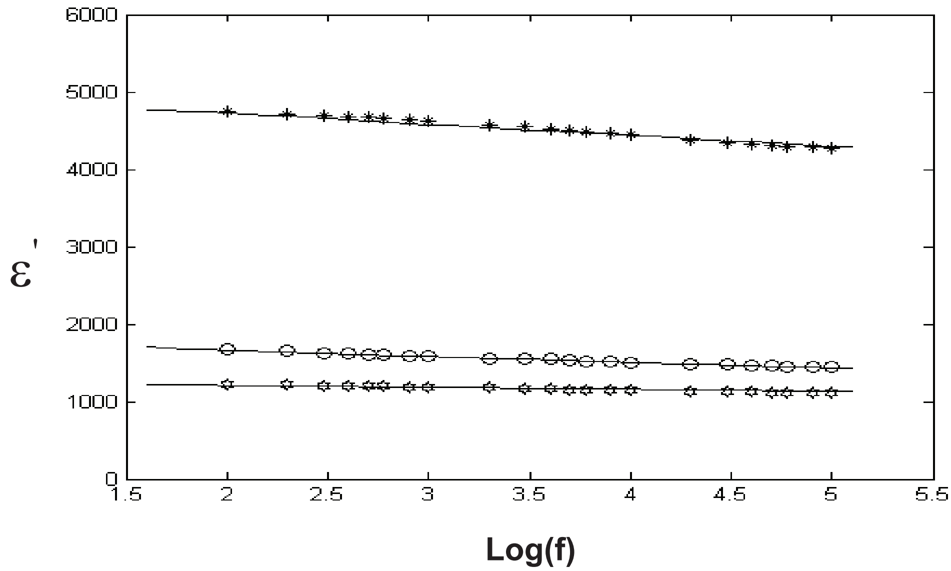

Fig.(3): comparison between computed and measured real part of the dielectric constant for CNO ceramics.

☆ : Experimental at 75K
O : Experimental at 126 K
✳ : Experimental at 180 K
___: Theoretical fits to two arcs

The procedure adopted to evaluate the arcs indicated strong interference between successive arcs. Thus, within the limited range of frequency reported one can not extract reliable dispersion parameter such as the relaxation time with subsequent evaluation of activation energy through Arrhenius relation. Therefore, measurements over a wider range of frequency are necessary prior to any accurate analysis. It is anticipated that such measurements will reveal most of the modes of relaxation in the material. Measurements under different temperatures will then provide means to identify the mechanisms underlying the modes. The limits in frequency range can simply be assessed from the fact that the lossy component of the dielectric constant ε'' diminishes at low and high frequencies.

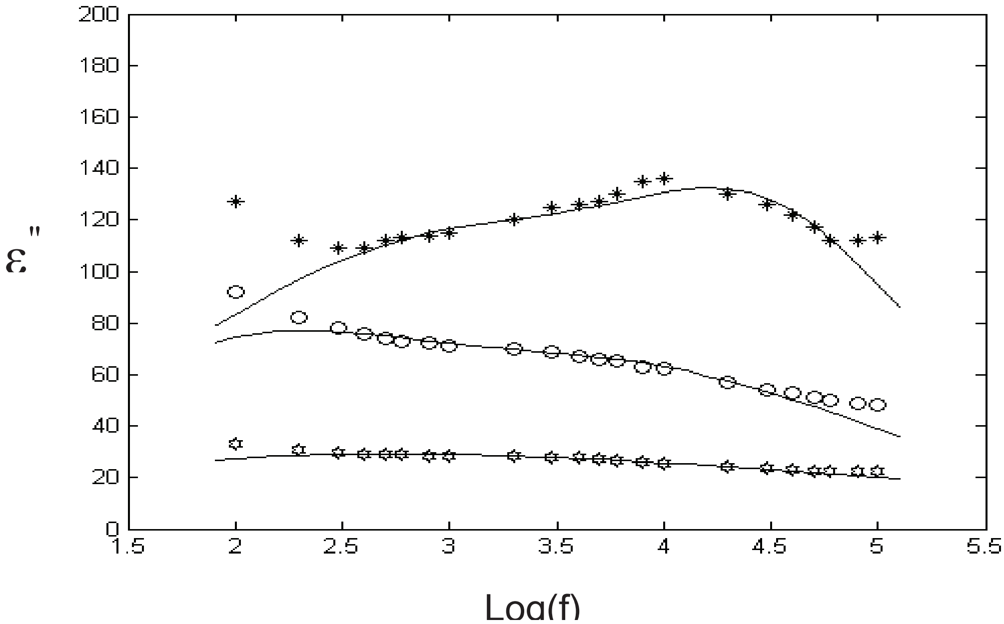

Fig.(4): comparison between computed and measured imaginary part of the dielectric constant for CNO ceramics.

☆: Experimental at 75K
O : Experimental at 126 K
✳ : Experimental at 180 K
___: Theoretical fits to two arcs

3. Conclusion

The multiple-arc approach adopted in this study has revealed a ferroelectric attribute in the CNO Ceramic through the relaxation spread parameters of the high frequency arcs. Meanwhile, the study has pointed out that those reported results concluded by part of the dispersion spectrum may not be strongly valid if the spread parameter α is relatively high. In such cases the collected dispersion data are mere superposition of various polarization effects, and so can not confidently be ascribed to a particular effect. However, employing the versatile multiple-arc analysis requires wider frequency coverage of dispersion.

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